



PATENT APPLICATION

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the Application of:) Paper No. 16
)
APPLICANTS: WILLIAM H. GONG,) Customer No. 04249
MONICA R. REGALBUTO &)
GEORGE A. HUFF JR.)
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)
SERIAL NO: 09/779,287) Group Art Unit:
) 1764
)
FILED: February 8, 2001) Examiner:
) Walter D. Griffin
)
)
FOR: PREPARATION OF COMPONENTS FOR) Attorney Docket
REFINERY BLENDING OF) No.: 37,248.04
TRANSPORTATION FUELS)

Commissioner for Patents
P. O. Box 1450
Alexandria VA 22313-1450

Response Under 37 CFR 1.116
-Expedited Procedure-
Examining Group

AFFIDAVIT UNDER 37 CFR § 1.132

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| <u>CERTIFICATE OF MAILING</u> | |
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| I hereby certify that this correspondence is being deposited with the United States Postal Service with sufficient postage as first class mail in an envelope addressed to: | |
| Commissioner of Patents and Trademarks, Alexandria VA 22313-1450 | |
| on <u>February 13</u> 2004 | By <u>Carol M. Neth</u> |
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| (Typed or Printed Name of Person Signing Certificate) | |

AFFIDAVIT UNDER 37 CFR § 1.132
of Dr. WILLIAM H. GONG.

5 1. I, WILLIAM H. GONG being duly sworn, depose and say:

 2. That I reside at 900 Fairfield Avenue, Elmhurst, Illinois
60126.

 3. In 1984 I received a Bachelor of Science degree in
Chemistry from the Illinois State University, Normal, Illinois, USA.
10 My undergraduate research was directed to methods for upgrade
of Illinois No. 6 coal.

 4. I received a Doctor of Philosophy Organic Chemistry in
1990 from the Iowa State University of Science and Technology,
Ames, Iowa, USA. My dissertation research was development of
15 palladium-based homogeneous catalytic methods and organic
synthesis.

 5. From 1990 to the present, I have been employed by
Amoco Chemical Company, now BP Chemicals, a corporation of the
State of Delaware, and have the position of Research Associate
20 Scientist. My responsibilities have included "step-out" research in
homogeneous catalytic oxygenation of alkylaromatic hydrocarbons,
and reaction mechanism, and development of processes to
catalytically oxygenate distillates and to selectively desulfurize
distillates. Among my present professional assignments are
25 Technical Manager of Innovation Portfolio Group responsible for
development of new catalytic oxidations processes; Technical
consultant in areas of catalytic oxidation chemistry and reaction
mechanism; and Manager of University Collaborations for the
technology department of business unit.

6. I am one inventor of the claimed subject matter of the above identified patent application.

7. I have read Primary Examiner Griffin's Office Action, mailed October 15, 2003, for our application and the references
5 relied upon to reject our claims. I wish to respond his objections.

8. The Examiner states that if one were to take a two-step process described in Hatanaka et al. (US 6,217,748) and combine it with the disclosure of Collins, Lucy, and Smith (EP 0482841 A1), and Ford et al. (US 3,341,448), one would arrive at our claimed
10 process.

9. U.S. Patent No 6,217,748 in the name of Hatanaka et al. teaches treatment of feeds a feedstock for diesel, catalytically hydrodesulfurize (HDS) it to some level, distill it to produce a lower boiling fraction that is substantially sulfur-free, and a higher
15 boiling fraction that is sulfur rich. Presumably, the sulfur impurities not removed in the first HDS step are the much more difficult varieties to remove (dimethyldibenzothiophenes). The high boiling fraction is then subjected to a second HDS, at more severe process conditions, to remove sulfur. The untreated lower
20 boiling fraction is then combined with the higher boiling, but treated fraction to produce a product.

10. Contrary to the position of Examiner, that our oxidative desulfurization (ODS) of a higher boiling fraction is equivalent to a second HDS, I argue that in fact our claimed process provides
25 substantial and unexpected improvements over teachings of Hatanaka et al. The final product of our claimed process is one that has superior properties to one that is produced by two HDS steps. Therefore, our process represents a patentable improvement, because this improvement was not expected even at the filing date
30 of our application.

11. At the time of our filing, there was a "general" assumption that sulfur containing organics, while unwanted, do impart a certain anti-wear properties. This "lubricity" characteristic is needed for efficient operation of a diesel engine.

5 The belief was that once the sulfur impurities were removed, for example via either ODS or HDS, lubricity additives must be added to the final product. This theory was borne out certainly in HDS treated diesel. However, one peer reviewed publication recently came to our attention, "The Lubricity of Diesel Fuels" (Wei, D.;
10 Spikes, H. A. *Wear*, **1986**, *111*, 217 - 235). In this journal article, the investigators reported that aromatic hydrocarbons, particularly polyaromatic hydrocarbons prevent wear or have lubricity characteristics. Furthermore, their investigation revealed that in fact sulfur-impurities are pro-wear.

15 12. The severe conditions that are required to do HDS for very low sulfur product have the unintended consequence of also hydrogenating polyaromatic hydrocarbons. This effect causes the disappearance of these valuable lubricity agents. Where the ODS step now has a superior advantage over the HDS process is that the
20 ODS process targets only the sulfur atom of these unwanted impurities. The mechanism of the oxidation is one that excludes any reactions with the polyaromatic hydrocarbons because there are no sulfur or nitrogen atoms (further discussion of nitrogen to be found later) in these polyaromatic hydrocarbons.

25 13. Our claimed process provides a substantial and unexpected improvements^{to} any HDS only process for the production of very low sulfur diesel and distillate products.

30 14. Collins et al. does not disclose or suggest that their process could impart the advantages of ODS over HDS as discussed above (i.e. preservation of anti-wear polyaromatic hydrocarbons). Their process did not take advantage of the effective strategy that our claimed process has of combining a preliminary HDS followed by distillation, and an ODS treatment of the higher boiling fraction.

15. The Examiner cites the Ford et al. reference of record as one that we could have borrowed from to combine with Hatanaka et al. and further with Collins et al. to result in our claimed process. Ford et al. teach that the degree of desulfurization is increased by combining both HDS and ODS. However, Ford et al. teaches that there is no importance to the order of HDS and ODS so as long as both steps are conducted. This is seen in the Ford reference (column 3, lines 18 - 20) where he states "This improvement is, moreover, independent of the order in which the stages are carried out."

16. By contrast, our claimed process has an advantageous strategy that Ford et al. disclose or suggest. Our claimed process has a strategy that permits optimization of both the HDS and the ODS processes. The use of hydrogen for an optimum level of desulfurization in the preliminary step permits one to also conduct the oxidation using precious hydrogen peroxide in the second step. The degree of desulfurization by HDS can change based on variable costs (i.e. cost of hydrogen). In cases where hydrogenation is less costly, perhaps more hydrogen can occur and less oxidation would be needed. The opposite can also apply.

17. Additionally, Ford et al. make no restriction on the type of feeds that can be desulfurized by their process using two HDS steps. The examples in Ford et al. use a Kuwait atmospheric residue, which contains weight percent levels of heavy metals and olefins. By contrast, our claimed process is applied to distillates that have already been processed, in particular a petroleum distillate consisting essentially of material boiling between about 50° C. and about 425° C. The presence of heavy metals interfere with the HDS and ODS. For example, the presence of heavy metals such as iron can catalyze unwanted Fenton reactions in the presence of hydrogen peroxide. A reference such as "Metal-Catalyzed Oxidations of Organic Compounds" by Sheldon and Kochi (Academic Press, New York, 1981, pp 35, 171, 177, and 330)

describes the mechanism of these reactions. A Fenton reaction would result in a rapid consumption of hydrogen peroxide and divert the process away from oxidizing sulfur impurities. The presence of many heavy metals will severely deactivate the HDS catalyst. Furthermore, HDS processes are not specific for sulfur atoms. The HDS catalyst employed will also target the hydrogenation/saturation of olefins, and ultimately consumes hydrogen. Beneficially in our claimed process, impurities such as heavy metals and much of the olefins are avoided so that precious hydrogen and hydrogen peroxide are used specifically to reduce levels of sulfur-containing and/or nitrogen-containing organic compounds.

18. The Examiner objects to our claims directed to a specific class of quaternary ammonium salts, because these salts are chemically similar to a phase transfer catalyst disclosed in Collins et al. Respectfully, I disagree with this position of Examiner. In fact, Collins et al. broadly claimed (Claim 6) "A process as claimed in claim 1 wherein step (1) is carried out in the presence of a phase transfer catalyst which is a quaternary ammonium or phosphonium salt."

19. In particular, our claim recites that the quaternary ammonium salt must at least have one of its alkyl (R) group be a methyl (CH₃) group. For those who are generally skilled in the art of phase transfer catalysis and organic chemistry, of which quaternary ammonium salts are generally known, not all quaternary ammonium salts can perform equally as well. The general formula of a quaternary ammonium salts have the following formula: R₄ N⁺ X⁻. By contrast, our claims recite that the soluble quaternary ammonium salt is represented by formula

CH₃ N (R)₃ X

where X is a halogen, sulfate, or bisulfate anion, and the R's are the same or different hydrocarbon moieties of at least 4 carbon atoms.

20. Examiner states that "one would expect the sulfur
5 compounds to be effectively oxidized regardless of the presence of
nitrogen." However, nitrogen is also oxidized along with sulfur by
hydrogen peroxide and the oxidation catalyst. The oxidation of
sulfur cannot occur exclusively in the presence of nitrogen. The
presence of nitrogen interferes with the sulfur oxidation by
10 consuming the oxidant. The oxidations of sulfur and nitrogen
atoms of organic compounds are generally known by those who are
skilled in the arts of oxidation reaction mechanisms. For example,
a source such as "Metal-Catalyzed Oxidations of Organic
Compounds" by Sheldon and Kochi (Academic Press, New York,
15 1981, pp 51, 54, and 229) clearly teaches that hydrogen peroxide
is an effective oxidant for these types of compounds. In order for
one to design an ODS process for a feedstock, one must be mindful
of the presence of nitrogen as it will consume the same oxidant.

21. The Examiner states that it would have been obvious to
20 one who is skilled in the arts of organic separation that a
combination of solvent extraction and adsorption would produce a
purer product than using one or the other process (i.e. extraction or
adsorption). Respectfully, I disagree with the Examiner. For those
who are generally skilled in the arts of analytical and organic
25 chemistries, the two processes cannot usually be linked together to
produce a purer product. Therefore, it is not obvious. For
example, solvent extraction usually precedes an adsorption
process. But, if the extraction unavoidably leaves behind traces of
solvent and the traces of solvent cannot be removed, then this
30 solvent will interfere with a subsequent adsorption process. Such
solvents include dimethyl sulfoxide, for example. When this
solvent enters an adsorption column, it will immediately occupy
the active sites of the adsorbent and immediately render the

adsorbent ineffective. In this case, one cannot combine both processes to produce a purer product..

22. I wish to present an example of where a solvent extraction followed by an adsorption process do not give higher performance. The example is for dimethyl sulfoxide, a solvent demonstrated as a extracting agent for sulfur impurities from a distillate. An article was published in a peer review journal (*Energy & Fuels* 2000, 14, 1232).

23. In these cases, a number of solvents were used to remove sulfur impurities. However, as with all extracting solvents, they do remain behind in the extracted materials. The advantages of these effective extracting solvents are also disadvantages in other ways. Their higher polarity than the distillate makes them immiscible, but their presence in the distillate also causes these trace levels of solvents to bind strongly with the adsorbent such as silica gel and alumina. By their strong binding to the active sites of the adsorbent, the trace solvents would render adsorbent ineffective for removing traces of oxidized sulfur impurities in the ODS process. Water as an extracting solvent is an exception because following any water extraction, the distillate can be treated separately with something such as molecular sieve or anhydrous inorganic compounds to easily remove water, *then* the distillate can be treated with an adsorbent to remove the traces of oxidized sulfur compounds in a polishing step.

24. On the other hand, if water were used in a liquid-liquid extraction, the extracted product can be readily dehydrated to remove traces of water, and then the product can be subjected to an adsorption process without interference from water. Because water is in fact used in the process, we can couple a solvent extraction process with an adsorption process to produce a product of superior purity.

25. Based on the above information, I respectfully argue with the Examiner that those who are ordinarily skilled in the art would not say that both the Hatanaka et al. HDS process and our novel process produce the same results because the results are in fact very different. The HDS process encounters severe challenges to achieve the ultra-low sulfur levels, which ~~the~~ our novel process does not encounter because the nature of the chemistry is vastly different. W4G 2/12/04

26. Therefore, I must contend that both processes produce substantially different result in terms of fuel properties, which are highly important. Our novel process clearly produced a product that has unexpectedly better properties than the HDS process because our process preserves the aromatics for lubricity. W4G 2/12/04

AND FURTHER AFFIANT SAYETH NOT.

15 William H. Gong
WILLIAM H. GONG

STATE OF ILLINOIS)
) SS.
COUNTY OF ~~DUPAGE~~ DEKALB ⁷⁶³

20 Sworn to and subscribed before me, a Notary Public, by said
WILLIAM H. GONG, on this 12 day of February 2004.



Karen G. Spangler
Notary Public